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Title: DETERMINATION OF THE INDEX OF REFRACTION FOR ALPHA-NTO AND DAAF USING THE BECKE TEST

Author(s): M. R. Oldenborg, DX-2
P.D. Peterson, DX-2
K-Y. Lee, DX-2

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Determination of the Index of Refraction for α-NTO and DAAF Using the Becke Test

M.R. Oldenborg, P.D. Peterson, K-Y. Lee
Los Alamos National Laboratory, Los Alamos, New Mexico, U.S.A.

ABSTRACT

NTO and DAAF are insensitive high explosives developed at Los Alamos National Laboratory. Both the defense and civilian sectors have particular interest in these types of materials for applications ranging from weapons to air bag design. The performance of explosives is highly dependent on particle size. Many common techniques for measuring particle size distributions require knowledge of the material's index of refraction. To-date the principle refractive indices of α-NTO and DAAF have not been determined. We present the three principle indices of refraction for the triclinic explosive α-NTO and an averaged index of refraction for DAAF found using the Becke Test. In addition, by comparing particle size distributions based on different refractive indices we show the importance of using the true index of refraction in measuring fine particles.

Keywords: Index of Refraction, NTO, DAAF, particle size

INTRODUCTION:

Developed at Los Alamos National Laboratory, NTO (5-nitro-2,4-dihydro-3H-1,2,4-triazole-5-one) and DAAF (4,4'-diamino-3,3'-azoxyfurazan) are both insensitive high explosives used by civilian and defense sectors. The performance of high explosives is highly dependent on particle size; therefore proper particle size measurement is key in predicting an explosive's properties and behavior. One method used to measure particle size is light scattering. Mie theory predicts the scattering angle of refracted light, given a spherical particle of known size, and refractive index. Therefore, particle size distributions (equivalent diameters) can be determined by measuring the scattering angle of light of a known wavelength, from particles with a known index of refraction. Knowledge of the compound's index of refraction is therefore paramount for obtaining accurate particle size distributions.

METHOD:

One effective method to determine a material's index of refraction is through a Becke Test. First, a small sample of the compound is immersed in an index of refraction matching fluid and prepared on a microscope slide. Using a polarized light microscope in a backlit configuration at magnifications ranging from 50X to 200X, a Beck Line is observed around the individual crystal. A Becke Line is a bright halo around the rim of a transparent particle that can be seen using a polarized light microscope. The Beck Line moves to the higher refractive index medium when the focus position is raised (focal distance shortened). Similarly, when the position of focus is lowered (increased focal distance) the Beck Line will move toward the lower refractive index medium. Using many different index of refraction fluids through an iterative process will ultimately yield the matching index of refraction. The accuracy of these measurements depends on the resolution of the index matching fluid set used. An accuracy of 0.005 index units was achieved for the index of refraction results presented in this paper.

RESULTS AND ANALYSIS:

Using the Becke line test, refractive indices were determined for α-NTO and DAAF; α-NTO is a triclinic crystal with three oblique indices of refraction (Figure 1). DAAF is monoclinic, and has three unique principle indices, two of which are orthogonal (Figure 2).

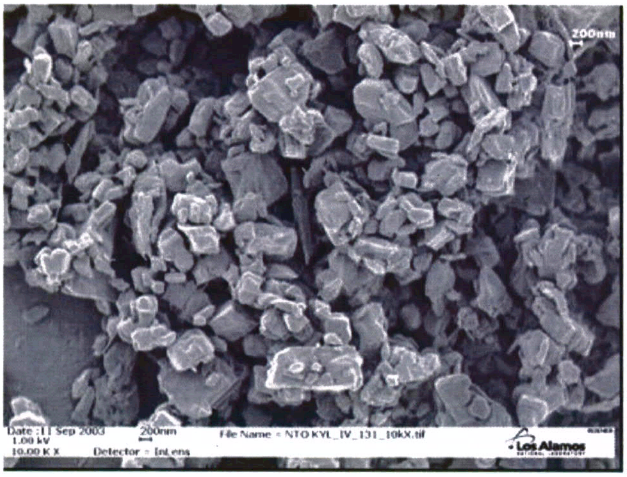

Figure 1: α-NTO at 10 kX Magnification (Triclinic Crystal)

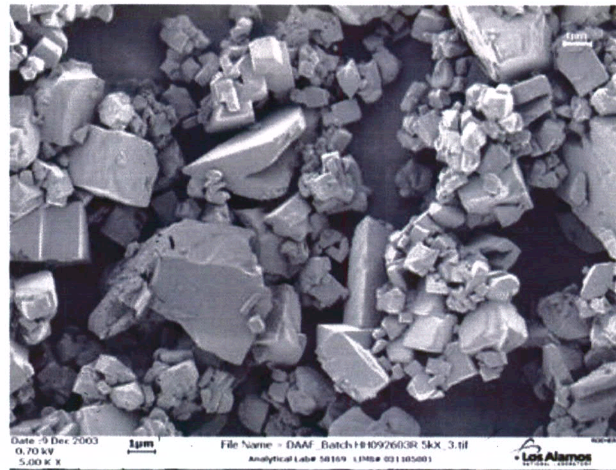

Figure 2: DAAF at 5 kX Magnification (Monoclinic Crystal)

Using the Becke line method, we have determined the principle refractive indices ($\eta_\alpha\eta_\beta\eta_\gamma$ for α-NTO and DAAF as shown in Table 1. Differences in the principle refractive indices for DAAF could not be resolved, so an average refractive index (η) for DAAF was assumed as shown in the table. Given a sufficient number of randomly oriented particles, this assumption is valid.

Table 1: α-NTO Indices of Refraction (Becke Test)

α-NTO	Index of Refraction
η_α	1.700
η_β	1.715
η_γ	1.720
DAAF	**Index of Refraction**
η_{avg}	1.830

Using the refractive index for DAAF as determined using the Becke test, the particle size distribution for a batch of DAAF (LANL Batch #HH092603R) was determined through light scattering using Beckman-Coulter LS 230. The LS 230 particle size analyzer uses the diffracted scattering angle of laser light (750 nm), to determine the size distribution of particles in a liquid suspension. 126 detectors measure the composite diffraction pattern. The pattern measured by the analyzer is the sum of the patterns scattered by each constituent particle in the sample and the reported measurement is the relative volume of spherical particles of that size.[1] The instrument is capable of measuring particle sizes from 0.04-2000 microns.

To prepare the suspension and attempt to break up any agglomerates in the sample prior to the analysis, a suspension of DAAF in DAAF-saturated water was sonicated for 1 minute in an ultrasonic bath, followed by the addition of 1 drop of 1% Triton X-100 (surfactant). Due to slight solubility of DAAF in water, the DAAF-saturated water was obtained by stirring excess amount of DAAF in water for 24 hours, followed by filtration. The resulting well-disbursed DAAF suspension was then analyzed for size distribution using the LS 230 particle analyzer.

The refractive index for DAAF determined by the Becke method was used in the analysis. The reported size distribution of DAAF is the average of three runs using a refractive index (R.I.) of 1.830. The measured size distributions of DAAF are illustrated in Figure 3. It can be seen that the majority of the particles in the sample volume fall in the range between 0.6 –15μm and there are small amounts of particles having sizes larger than 35 μm. SEM images of the same batch of DAAF further verify the measured size distribution (Figures 4-5). In addition, the measured size distributions of DAAF as a function of assumed refractive index are shown in Figure 6. These results clearly show the effect of using an incorrect, assumed refractive index on measured particle size distributions.

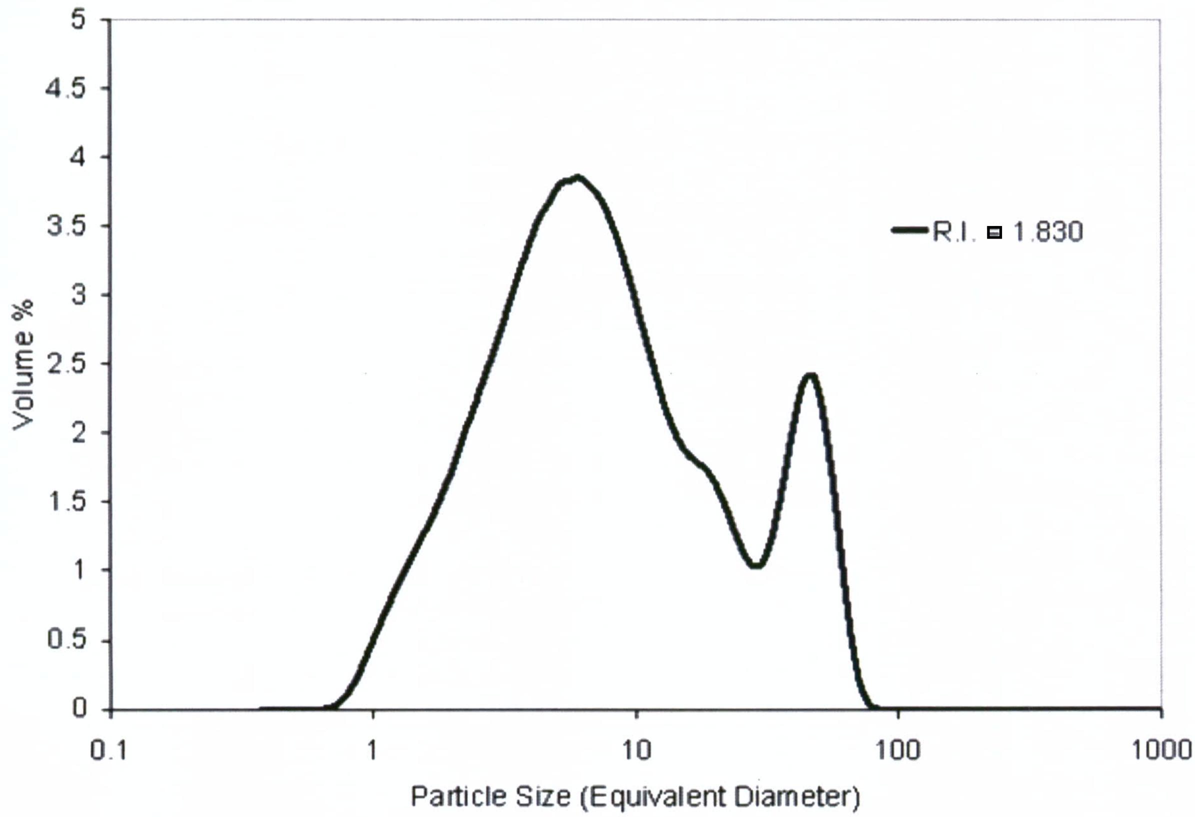

Figure 3: DAAF Particle Size Distribution (Index of refraction = 1.830)

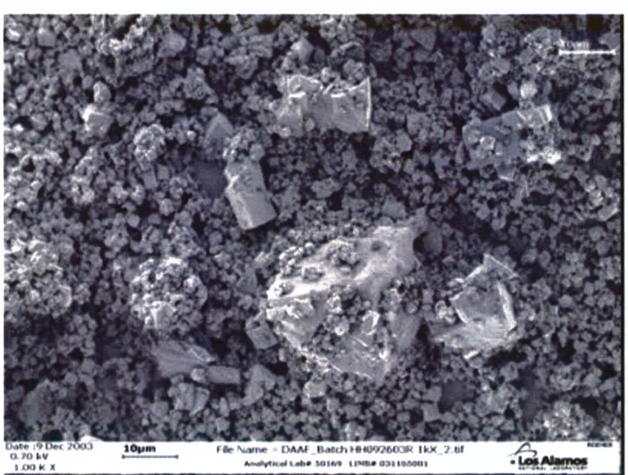

Figure 4: DAAF (1000 X Magnification)

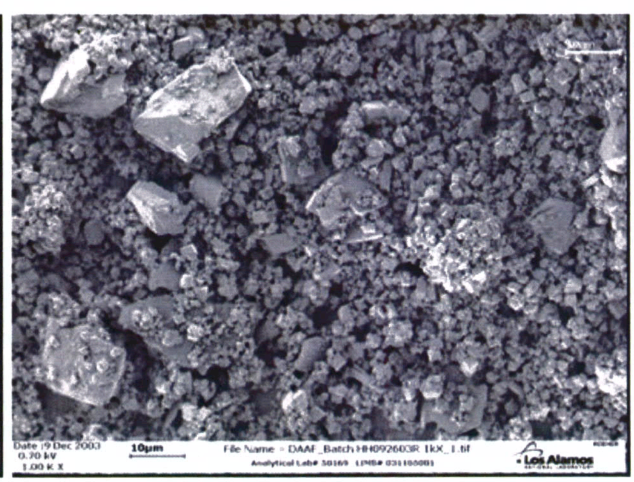

Figure 5: DAAF (1000 X Magnification)

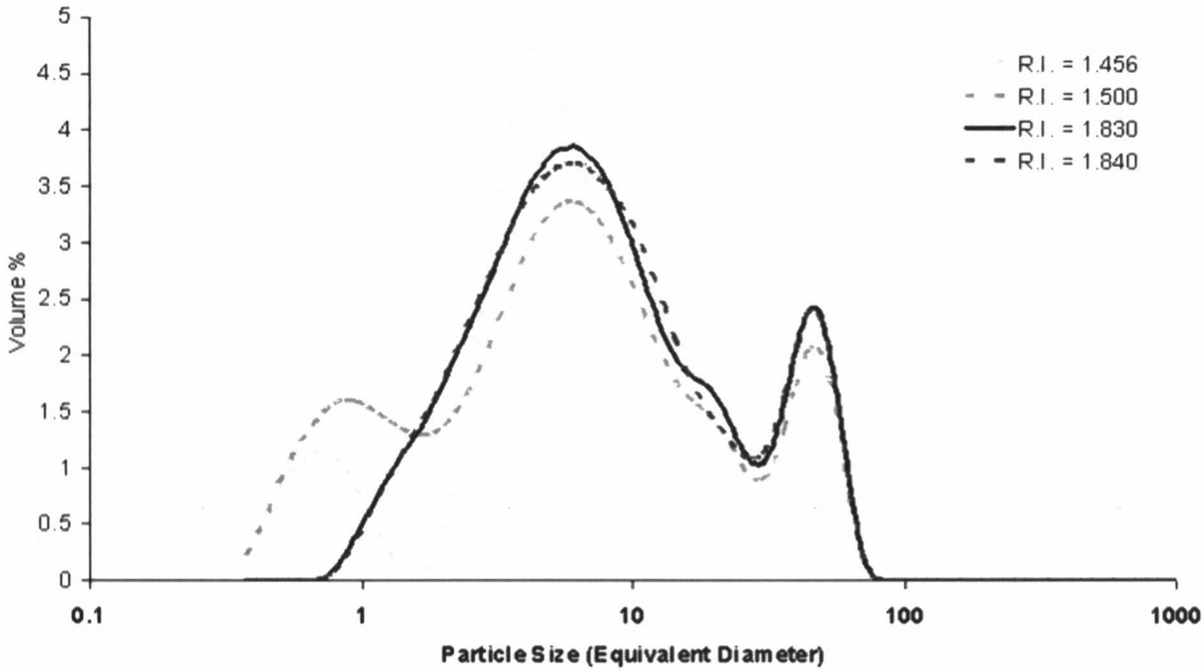

Figure 6: LS 230 Particle Size Results for DAAF Using Different Refractive Indices (R.I.) (True Index of Refraction for DAAF = 1.830)

CONCLUSION:

Particle size has been shown to greatly affect an explosives performance. Therefore, accurate measurements of an explosives particle size are desired. The Beckman-Coulter LS 230 particle size analyzer and other similar modules require the knowledge of the explosive's index of refraction. Using the Becke Test the indices of refractions for both α-NTO and DAAF were determined and their results were published for the first time in this paper. Tests were conducted to quantitatively measure the effect of inaccurate index of refraction fluids using the Beckman-Coulter LS 230 particle size analyzer. These results show that the proper index of refraction must be used for proper particle size measurements.

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[1] Product Manual, Coulter LS Series, Beckman-Coulter, Inc.